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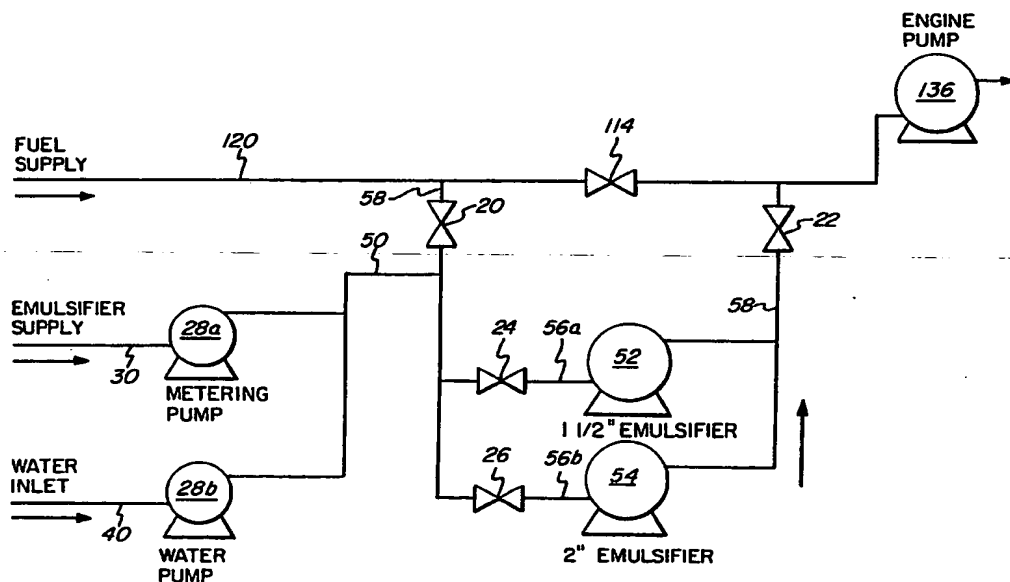
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(54) Title: PROCESS FOR REDUCING NITROGEN OXIDES EMISSIONS AND IMPROVING THE COMBUSTION EFFICIENCY OF A TURBINE



(57) Abstract

A process for formation of an emulsion in emulsifiers (52 and 54) with water from water inlet (40) and fuel from fuel supply (120) and optionally an emulsifier from emulsifier supply (30) and injection of said emulsion into a combustion zone of a turbine by engine pump (136) to reduce nitrogen oxides and plume opacity and improve the combustion efficiency of a turbine.

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DESCRIPTION**PROCESS FOR REDUCING NITROGEN OXIDES EMISSIONS AND
IMPROVING THE COMBUSTION EFFICIENCY OF A TURBINE**Related Application

This application is a continuation-in-part of copending and commonly assigned U.S. Patent Application entitled "Improved Combustion Efficiency Water-in-Fuel Oil Emulsion", having Serial No. 07/603,266, filed in the name of Sprague on October 24, 1990, which in turn is a continuation of U.S. Patent Application Serial No. 07/348,296, filed May 5, 1989, now abandoned, the disclosures of which are incorporated herein by reference.

Technical Field

The present invention relates to a process which will improve the combustion efficiency of a gas turbine in order to reduce the emissions of nitrogen oxides (NO_x) and visible emissions (particulates, which lead to plume opacity) to the atmosphere.

Gas or combustion turbines have been utilized by many utilities as peaking units to rapidly bring additional electrical generation on line as required and, hence, are preferred for many applications. Unfortunately the

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temperatures at which gas turbines operate tend to cause the production of thermal NO_x , the temperatures being so high that free radicals of oxygen and nitrogen are formed and chemically combine as nitrogen oxides. Nitrogen oxides are troublesome pollutants and comprise a major irritant in smog. It is further believed that nitrogen oxides can cause or enhance the process known as photochemical smog formation through a series of reactions in the presence of sunlight and hydrocarbons.

Moreover, nitrogen oxides are a significant contributor to acid rain and have been implicated in the undesirable warming of the atmosphere through what is known as the "greenhouse effect" and in the depletion of the ozone layer. In addition, gas turbines often emit a visible plume, which is highly undesirable since it causes concern among the population in areas surrounding the facility.

Although the use of emulsified oils (primarily produced from relatively simple mechanical techniques) for combustion improvements has been suggested in the past, they have not been applied to gas turbines, and the oils emulsified are usually heavy oils (i.e., #6 oil). Because gas turbines are peaking units, the fuel is required to remain emulsified for at least 30 days in a holding tank, and at least 2 hours during in-line mixing. This has been difficult to accomplish, especially when using mechanical emulsion methodology. In addition, gas turbines are very sensitive to corrosion, which often leads the practitioner to avoid introducing emulsified water into the combustion zone.

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It has been known for some time that the injection of water directly into the combustion zone (also referred to as the "combustion can") of a gas turbine at water to fuel ratios of greater than 1:1 can control the emission of NO_x and reduce opacity to a limited extent. The direct injection of water into the combustion can, though, requires extensive mechanical modification of the gas turbine (with high capital cost) and involves the injection of large volumes of demineralized water which results in extensive additional maintenance and outage time because of the thermal shock to the combustion can.

What is desired, therefore, is a process which permits the reduction of effluent nitrogen oxides and plume opacity from a gas turbine without the thermal shock, large capital costs, and other drawbacks of injecting water directly into the combustion can.

Brief Description of the Drawings

The present invention will be better understood and its advantages more apparent in view of the following detailed description, especially when read with reference to the appended drawings, wherein:

FIGURE 1 is a schematic illustration of a gas turbine fuel supply system having an emulsification system according to the present invention installed therein;

FIGURE 2 is a schematic illustration of an emulsification system according to the present invention as installed in a gas turbine fuel supply system; and

FIGURE 3 is a graphic representation of the results of Examples IIa and IIb.

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Disclosure of Invention

The present invention relates to a method for reducing nitrogen oxides emissions and improving the combustion efficiency of a gas turbine (which term will be considered to be interchangeable with combustion turbine). In particular, this invention relates to a process involving the formation of a stable water-and-fuel oil emulsion, where the oil is a light fuel oil such as diesel fuel, distillate fuel or #2 oil. The subject emulsion can be either a water-in-fuel oil or a fuel oil-in-water emulsion (although water-in-fuel oil emulsions are preferred for most applications), and the introduction of the emulsion into at least one of the combustion cans of a gas turbine through its fuel system.

Typically, the oil phase in the inventive emulsions comprise what is conventionally known as diesel fuel, distillate fuel, or #2 oil, as defined by the American Society of Testing and Measurement (ASTM) Standard Specification for Fuel Oils (Designation: D 396-86). Especially preferred are distillate fuels. Included among these are kerosene and jet fuels, both commercial and military, commonly referred to as JP4 and JP5, respectively.

Although demineralized water is not required for the successful control of nitrogen oxides and opacity, the use of demineralized water in the emulsion formed according to the process of this invention is preferred in order to avoid the deposit of minerals from the water on the blades and other internal surfaces of the gas turbine. In this way, turbine life is extended and maintenance and outage time significantly reduced.

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The emulsions used in the fuel system of the gas turbine advantageously comprise water-in-fuel oil emulsions having up to about 50% water by weight. The emulsions of this type which have the most practical significance in combustion applications are those having at least about 5% water and are preferably about 10% to about 35% water-in-fuel oil by weight. In addition, this invention also relates to the formation of fuel oil-in-water emulsions having about 50% to about 80% water, which have practical applicability in certain situations.

Advantageously, the emulsions are prepared such that the discontinuous phase (i.e., the water in a water-in-fuel-oil emulsion and the oil in an a fuel oil-in-water emulsion) has a particle size wherein at least about 70% of the droplets are below about 5 microns Sauter mean diameter. More preferably, at least about 85%, and most preferably at least about 90%, are below about 5 microns Sauter mean diameter.

Emulsion stability is largely related to droplet size. The primary driving force for emulsion separation is the large energy associated with placing oil molecules in close proximity to water molecules in the form of small droplets. Emulsion breakdown is controlled by how quickly droplets coalesce. Emulsion stability can be enhanced by the use of surfactants and the like, which act as emulsifiers or emulsion stabilizers. These generally work by forming repulsive layers between droplets prohibiting coalescence. The gravitational driving force for phase separation is much more prominent for large droplets, so emulsions containing large droplets separate most rapidly.

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Smaller droplets also settle, but can be less prone to coalescence, which is the cause of creaming. If droplets are sufficiently small, the force of gravity acting on the droplet is small compared to thermal fluctuations or subtle mechanical agitation forces. In this case the emulsion can become stable almost indefinitely, although given a long enough period of time or a combination of thermal fluctuations these emulsions will eventually separate.

Because of the operating characteristics of gas turbines, it is required that the water/fuel oil emulsion exhibit a high degree of stability. In most cases, gas turbines are "peaking" units, as noted, which do not operate regularly. Accordingly, an emulsified fuel may sit stagnant for extended periods or with only mild recirculation in the fuel line. In order to avoid separation of the emulsion into its components, which can cause slugs of water to be injected through the burner nozzle leading to combustion problems and possible engine damage, an emulsifier or emulsion stabilizer is also desirable in the water/fuel oil emulsion.

Advantageously, the emulsifier utilized comprises a ~~composition selected from one or more alkanolamides, by~~ which is generally meant an amide formed by condensation of an alkyl or hydroxyalkyl amine or mixtures thereof, and an organic acid. Preferred acids are fatty acids, such as lauric acid, linoleic acid, oleic acid, stearic acid, and coconut oil fatty acids. Most preferred are alkanolamides having a molar ratio of alkanolamine group to acid group of from about 1:1 to about 2:1.

Surprisingly, these compositions can stabilize an emulsion of up to about 50% water-in-fuel oil, or up to

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about 80% fuel oil-in-water in alkanolamide amounts as low as about 0.05% by weight, and even as low as about 0.01% by weight. In fact, although there is no true maximum amount of emulsifier which can be used, there is usually no need for greater than about 1%, or, in fact, greater than about 0.5% by weight emulsifier in the subject emulsion. Advantageously, to stabilize an emulsion of up to about 50% water-in-fuel oil, the noted alkanolamides should be included in an amount of from about 0.1% to about 0.3% by weight.

Suitable alkanolamides which can function to stabilize the emulsion of the process of the present invention include any one or more of the following: cocamide diethanolamine (DEA), lauramide DEA, polyoxyethylene (POE) cocamide, cocamide monoethanolamide (MEA), POE lauramide DEA, oleamide DEA, linoleamide DEA, and stearamide MEA, as well as mixtures thereof. Such alkanolamides are commercially available under trade names such as Clindrol 100-0, from Clintwood Chemical Company of Chicago, Illinois; Schercomid ODA, from Scher Chemicals, Inc. of Clifton, New Jersey; Schercomid SO-A, also from Scher Chemicals, Inc.; and Mazamide®, and the Mazamide series from PPG-Mazer Products Corp. of Gurnee, Illinois.

Other emulsifiers which may be useful include ethoxylated alkylphenols, such as nonyl phenol, octyl phenol, etc. and salts of alkylated sulfates or sulfonates, such as sodium lauryl sulfate. In addition, the skilled artisan will recognize that other emulsifiers or blends of emulsifiers, may be also effective at maintaining the stability of the inventive emulsion.

The use of the noted emulsifiers provides chemical

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emulsification, which is dependent on hydrophylic-lipophylic balance (HLB), as well as on the chemical nature of the emulsifier. The HLB of an emulsifier is an expression of the balance of the size and strength of the hydrophylic and the lipophylic groups of the composition. The HLB, which was developed as a guide to emulsifiers by ICI Americas, Inc. of Wilmington, Delaware can be determined in a number of ways, most conveniently for the purposes of this invention by the solubility or dispersibility characteristics of the emulsifier in water, from no dispersibility (HLB range of 1-4) to clear solution (HLB range of 13 or greater). The emulsifiers useful in the present invention should most preferably have an HLB of 8 or less, meaning that after vigorous agitation they form a milky dispersion in water (HLB range of 6-8), poor dispersion in water (HLB range of 4-6), or show no dispersability in water (HLB range of less than 4).

It is also possible to utilize a physical emulsion stabilizer in combination with the chemical emulsifiers noted above to maximize the stability of the emulsion achieved in the process of the present invention. Use of physical stabilizers also provides economic benefits due to their relatively low cost. Although not wishing to be bound by any theory, it is believed that physical stabilizers increase emulsion stability either by increasing the solubility of immiscible phases or by forming an insoluble barrier attracted to the oil/water interface. Exemplary of suitable physical stabilizers are waxes, cellulose products and gums such as whalen gum and xanthan gum.

When utilizing both chemical emulsifiers and physical emulsion stabilizers, the physical stabilizer is present

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in an amount of about 0.05% to about 5% by weight of the combination of chemical emulsifier and the physical stabilizer. The resulting combination emulsifier/stabilizer can then be used at the same levels noted above for the use of emulsifier alone.

The emulsification provided must be sufficient to maintain the emulsion to a greater extent than if the emulsifier was not present and to as great an extent as possible. The actual level of emulsification will vary depending upon the percentage of oil and water in the emulsion and the particular fuel oil utilized. For example, when the continuous phase is #2 oil, it is highly desired that no more than about 0.1% free water be present in the emulsion, and that the emulsion is maintained that way at ambient conditions for at least about two hours. Ambient conditions, that is, the conditions to which the emulsion is expected to be exposed, include the temperature in the gas turbine fuel feed lines. Such temperatures can be up to about 65°C, more typically up to about 90°C and even as high as about 100°C.

The emulsion used in the process of the present invention can be formed using a suitable mechanical emulsifying apparatus which would be familiar to the skilled artisan. Advantageously, the apparatus is an in-line emulsifying device for most efficiency. The emulsion is formed by feeding both the water and the fuel oil in the desired proportions to the emulsifying apparatus, and emulsifier or stabilizer when used can either be admixed or dispersed into one or both of the components before emulsification or can be added to the emulsion after it is formed.

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Preferably, the emulsifier and/or stabilizer is present at the time of emulsifying the water and fuel oil. Most advantageously, any emulsifier or stabilizer used is provided in the water phase, depending on its HLB. It has been found that the emulsions noted above with the chemical emulsifiers can be stabilized at up to about 50% water-in-fuel oil for up to 30 days and longer. In fact, with mild agitation, such as recirculation, it is believed that the emulsion can stay in suspension indefinitely.

Surprisingly, the emulsion can then be introduced into a combustion can of the gas turbine through the fuel feed lines and burner nozzles conventionally used with such combustion apparatus. There is no need for modification of the gas turbine fuel feed lines or combustion can to accommodate the emulsion used in the process of this invention.

Figures 1 and 2 illustrate a gas turbine fuel supply system having installed therein an emulsification system for the practice of the process of the present invention and a schematic illustration of the emulsification system itself. As illustrated in Figure 1, an emulsification system 10 can be installed in a gas turbine fuel supply system 100 between the heater 122 and the final filter 124. Although emulsification system 10 is illustrated as being installed in this position in fuel supply system 100, it will be recognized by the skilled artisan that other positions may be more advantageous in terms of emulsion stability in other fuel supply system embodiments, and emulsification system 10 can be installed at virtually any point along fuel supply system 100 for operability. Indeed, it will also be recognized that heater 122 and final filter 124 are preferred

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components of fuel supply system 100 and conventionally utilized, but not critically needed.

Fuel supply system 100 is typical of many gas turbine fuel supply systems and generally comprises a fuel supply line 110 which is fed by a fuel tank or other holding or storage apparatus (not shown). Fuel flowing through fuel supply line 110 proceeds through a set of initial filters 112a and 112b, and is then fed to individual fuel supply systems 120, 220, and 320 which feed engines controlled by fuel supply system 100. For ease of understanding, fuel supply system 120 which feeds engine manifold 130 is specifically illustrated. Supply systems 220 and 320 are equivalent in operation.

Fuel supplied through fuel supply line 110 is fed along engine manifold 130 supply line 120 into heater 122. From there, the fuel flow continues past valve 114 into final filter 124. From final filter 124, the fuel flow continues along line 120 through engine pump 136 and from there into fuel distribution manifold 121 which then supplies the fuel through primary nozzle 132 and secondary nozzle 134 to engine manifold 130, which is the combustion zone of the subject gas turbine. In addition, fuel supply system 110 further comprises recirculation lines 123a and 123b and recirculation pump 128 for recirculation of the fuel through line 120.

When valve 114 in fuel supply line 120 is closed and valves 20 and 22 in emulsification system 10 are open, fuel flowing along fuel supply line 120 is shunted through emulsification system 10 after heater 122, and is resupplied to fuel supply line 120 before final filter 124 for feeding to engine manifold 130 or recirculation.

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As illustrated in Figure 2, emulsification system 10 comprises an emulsifier supply line 30 which supplies emulsifier from a tank or other storage means (not shown) to a metering pump, and is then fed through line 50. In addition, emulsification system 10 comprises water inlet line 40 which feeds water from a tank or other supply means (not shown) through a water pump 28a to supply line 50 where it is admixed with emulsifier supplied from emulsifier supply line 30.

The water/emulsifier fed through line 50 then meets fuel being fed through line 58 when valve 20 is open and valve 114 is closed. These are then fed through either one or both of 1 1/2 inch emulsifier 52 or 2 inch emulsifier 54, depending on whether one or both of valves 24 or 26 is open through feed lines 56a and 56b, respectively. The emulsified water-in-fuel oil is then fed via line 58 back through fuel supply line 120 when valve 22 is open and from there into engine pump 136 and into engine manifold 130.

Although not wishing to be bound by any theory, it is believed that the use of an emulsion provides striking advantages over separate water injection systems because the water is being provided internal to the flame. By doing so, less water is required to achieve superior results, which reduces the deleterious effects of directly introducing large amounts of water to the combustion zone of the gas turbine.

Because of the advantages of introducing water internal to the flame, utilization of the inventive process results in a reduced use of demineralized water (since the emulsion contains less than the 1:1 ratio of water to fuel oil used when water is injected directly

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into the combustion can), and leads to less thermal stress which reduces maintenance cost and outage time.

When the emulsified fuel is introduced into the combustion zone, the heat of vaporization from the burning fuel causes the emulsified water droplets to become steam, which creates a secondary atomization. This secondary atomization improves combustion and increases the gas volume. In addition, the heat required to change the water to steam is believed to reduce the flame temperature of the combustion which helps to reduce formation of nitrogen oxides.

Additionally, use of the water/fuel oil emulsion results in substantial elimination of the need for an expensive, independent smoke suppressant additive. Typically, such additives are heavy metal based products which can form deposits on the turbine blades, reducing efficiency and increasing maintenance costs. By the use of emulsions in the process of this invention, a 90% or greater reduction in smoke suppressant additive use is often achieved, which increases the blade life due to reduced deposits, and creates less wear on the turbine blade coatings. These advantages all lead to significant savings in operating and maintenance costs.

Furthermore, when compared to a separate water injection system, the use of the process of this invention leads to improved engine fuel system integrity; the engine burns cooler, which, as noted, leads to less thermal stress; it is believed that the gas turbine can assume a higher load capacity; and compliance with environmental regulations is more easily obtainable.

When the process of the present invention is, for

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instance, conducted on a 48 megawatt gas turbine by installing a manually operated in-line emulsification system at the fuel oil inlet, it is found that reductions of nitrogen oxides of about 75%, which are immediate and reproducible, are obtained. These results are about 50% greater than those found when a separate water injection system is used at equivalent water injection rates. In addition, plume opacity is found to disappear and no operational problems are detected. In fact, inspection of the engine after use of the emulsion process of this invention shows no deposits.

The following examples further illustrate and explain the invention.

EXAMPLE I

An emulsification system is prepared comprising two rotary emulsifiers and related storage, pumping and piping apparatus for preparation and supply of a water-in-fuel oil emulsion to a Pratt and Whitney Jet engine burning 30 gallons of fuel per minute at full load (21MW).

Baseline emissions tests are run on the engine with non-emulsified distillate fuel oil, and then with emulsified fuel at water levels of 10%, 15%, 20%, 25%, 35%, and 50%. The emulsifier used is oleamide DEA added at 2.5 gallons per 1,000 gallons of fuel (corresponding to .25% of emulsifier by weight). The emulsion remained stable (i.e., no visible water separation) for over two hours without agitation.

The results of the two tests are then compared and the following found:

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1. When compared to baseline, each incremental increase in water content reduced nitrogen oxides levels up to 80%.

2. At water contents above 20%, visible opacity disappeared.

3. At water levels above 35%, power output from the engine increased by approximately 3% due to greater mass flow.

4. Blades and guide vanes are found to be cleaner with the emulsion prepared according to the present invention.

EXAMPLE IIa

An emulsification system in accordance with Figures 1 and 2 is prepared for supply to a single TP&M A4 engine operating as part of a twinpack rated at approximately 35 MW. Flue gas samples are obtained through a three point probe installed on the outlet duct with the sample points located between the guide vanes. The samples are combined and the NO and NO₂ levels therein measured, and compared with baseline levels.

Two tests are run using incrementally increased emulsion strengths (water content) and the results plotted in Figure 3.

EXAMPLE IIb

As reported by Becker et al. in "Gas Turbine Operating Performance and Considerations for Combined Cycle Conversion at Hay Road Power Station", American

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Power Conference, April, 1990, two 100 MW Siemens model V84.2 engines with hybrid burners are operated with separate water injection. Flue gas NO and NO₂ levels are measured and compared with baseline levels.

Two tests are run using incrementally increased levels of water injection and the results plotted in Figure 3.

Figure 3 illustrates the fact that use of the process of the present invention permits equivalent reduction of nitrogen oxides with approximately 50% of the amount of water injected.

The above description is for the purpose of teaching the person of ordinary skill in the art how to practice the present invention, and it is not intended to detail all of those obvious modifications and variations of it which will become apparent to the skilled worker upon reading the description. It is intended, however, that all such obvious modifications and variations be included within the scope of the present invention which is defined by the following claims.

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Claims

1. A process for reducing nitrogen oxides emissions and improving the efficiency of a gas turbine comprising forming an emulsion which comprises water and fuel oil and using said emulsion in the fuel system of a gas turbine.
2. The process of claim 1 wherein said emulsion comprises a water-in-fuel oil emulsion.
3. The process of claim 2 wherein said emulsion comprises up to about 50% water-in-fuel oil.
4. The process of claim 1 wherein said fuel oil is selected from the group consisting of distillate fuel, kerosene, jet fuel, diesel fuel, and No. 2 oil.
5. The process of claim 3 wherein said emulsion further comprises an emulsifier having an HLB of 8 or less in an amount of about 0.01% to about 1.0% by weight.
6. The process of claim 5 wherein said emulsifier comprises ethoxylated alkylphenols, alkylated sulfates, alkylated sulfonates, or alkanolamides formed by condensation of an alkyl or hydroxyalkyl amine or mixtures thereof and an acid.
7. The process of claim 6 wherein said alkanolamide emulsifier is selected from the group consisting of cocamide DEA, lauramide DEA, propoxylated cocamide MEA, cocamide MEA, propoxylated lauramide DEA, oleamide DEA, linoleamide DEA, stearamide DEA, and mixtures thereof.
8. The process of claim 7 wherein said alkanolamide

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emulsifier is present in an amount of about 0.05% to about 0.3% by weight.

9. The process of claim 6 wherein said emulsifier further comprises an emulsion stabilizer selected from the group consisting of waxes, cellulose products, gums, and mixtures thereof.

10. The process of claim 1 wherein said emulsion comprises a fuel oil-in-water emulsion.

11. The process of claim 10 wherein said emulsion comprises about 50% to about 80% water.

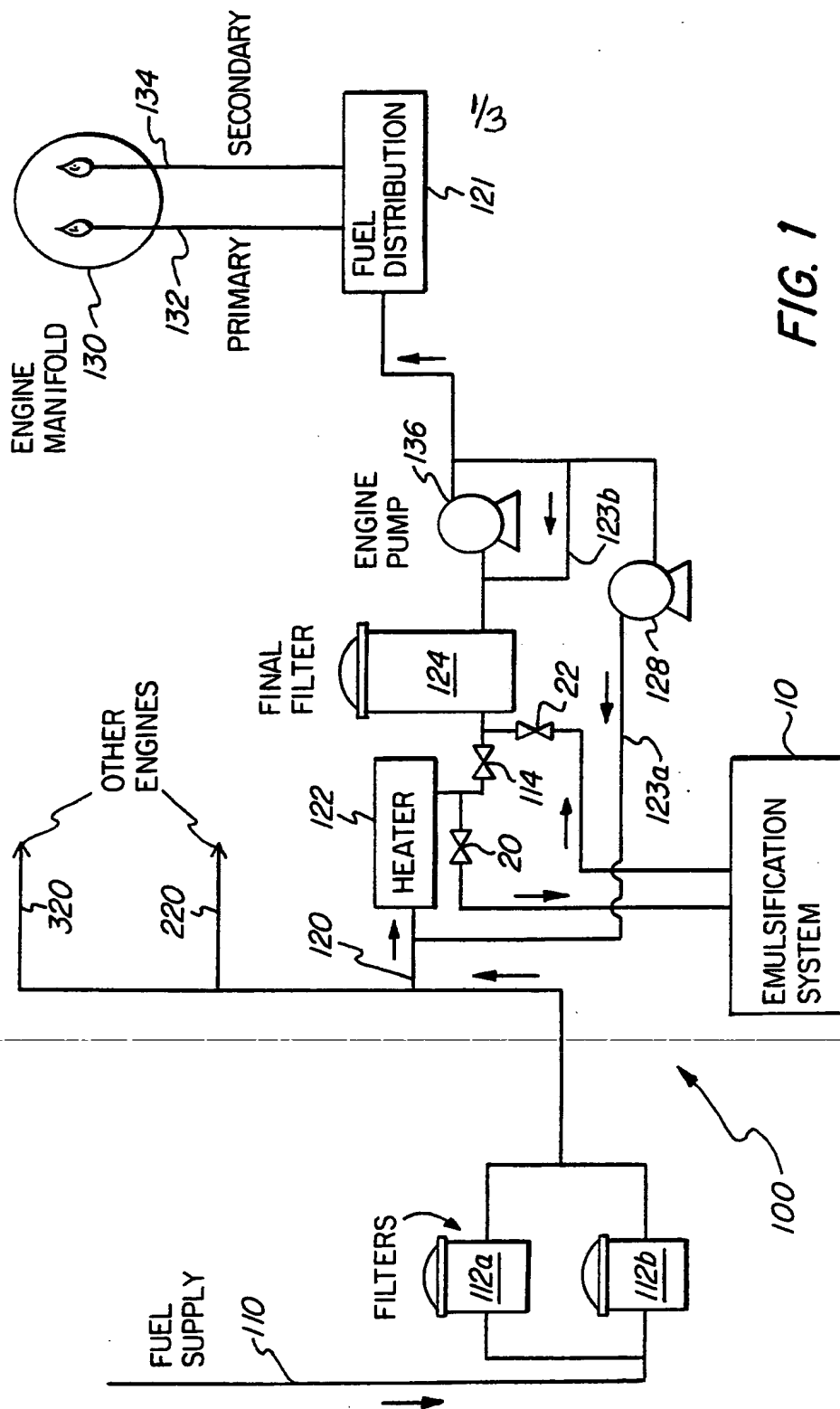
12. The process of claim 10 wherein said emulsion further comprises an emulsifier having an HLB of 8 or less in an amount of about 0.01% to about 1.0% by weight.

13. The process of claim 12 wherein said emulsifier comprises an alkanolamide formed by condensation of an alkyl or hydroxyalkyl amine or mixtures thereof and an acid.

14. The process of claim 13 wherein said alkanolamide emulsifier is selected from the group consisting of cocamide DEA, lauramide DEA, propoxylated cocamide MEA, cocamide MEA, propoxylated lauramide DEA, oleamide DEA, linoleamide DEA, stearamide DEA, and mixtures thereof.

15. The process of claim 14 wherein said alkanolamide emulsifier is present in an amount of about 0.05% to about 0.3% by weight.

16. The process of claim 13 wherein said emulsifier further comprises an emulsion stabilizer selected from the group consisting of waxes, cellulose products, gums, and mixtures thereof.



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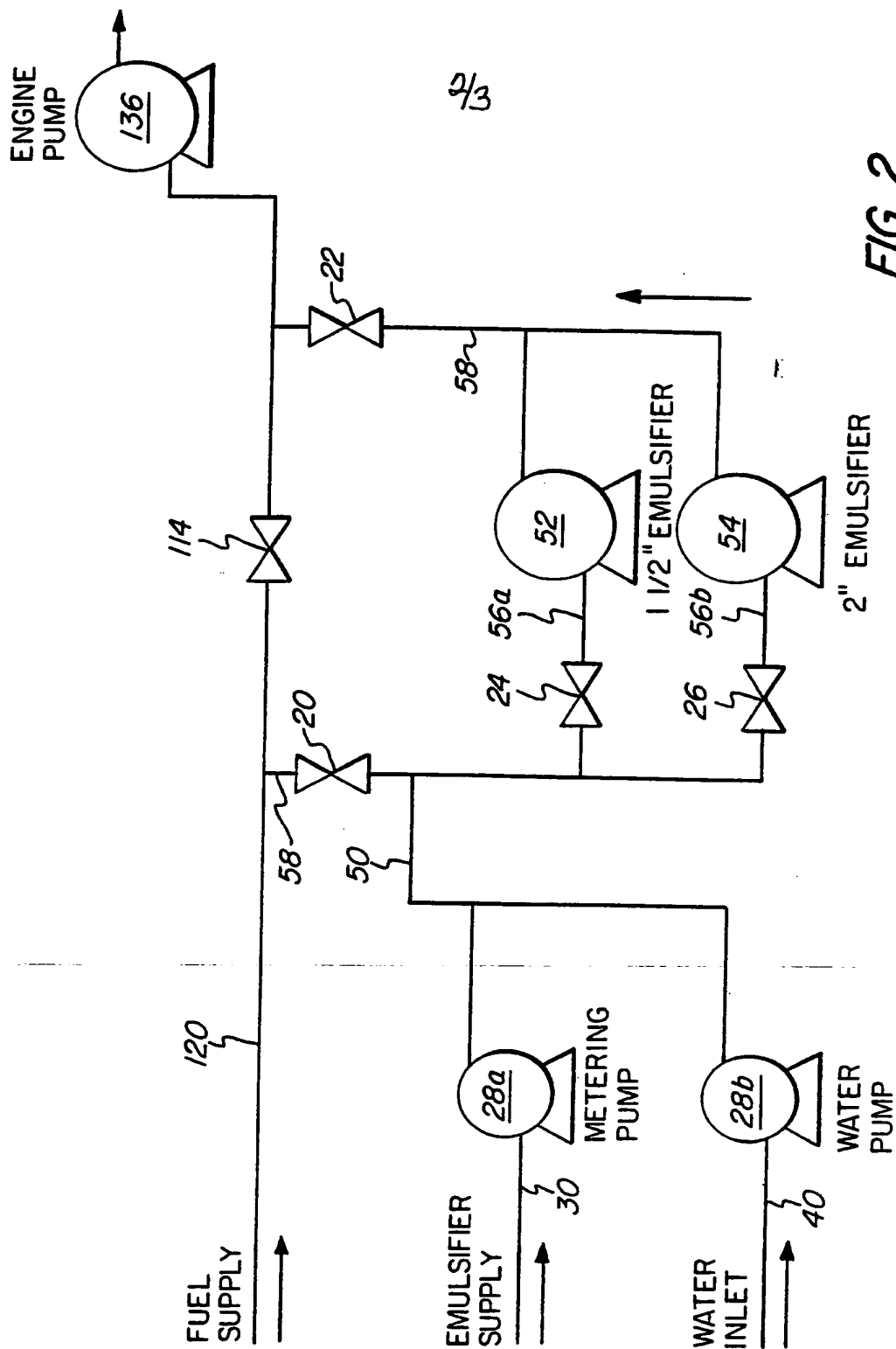


FIG. 2

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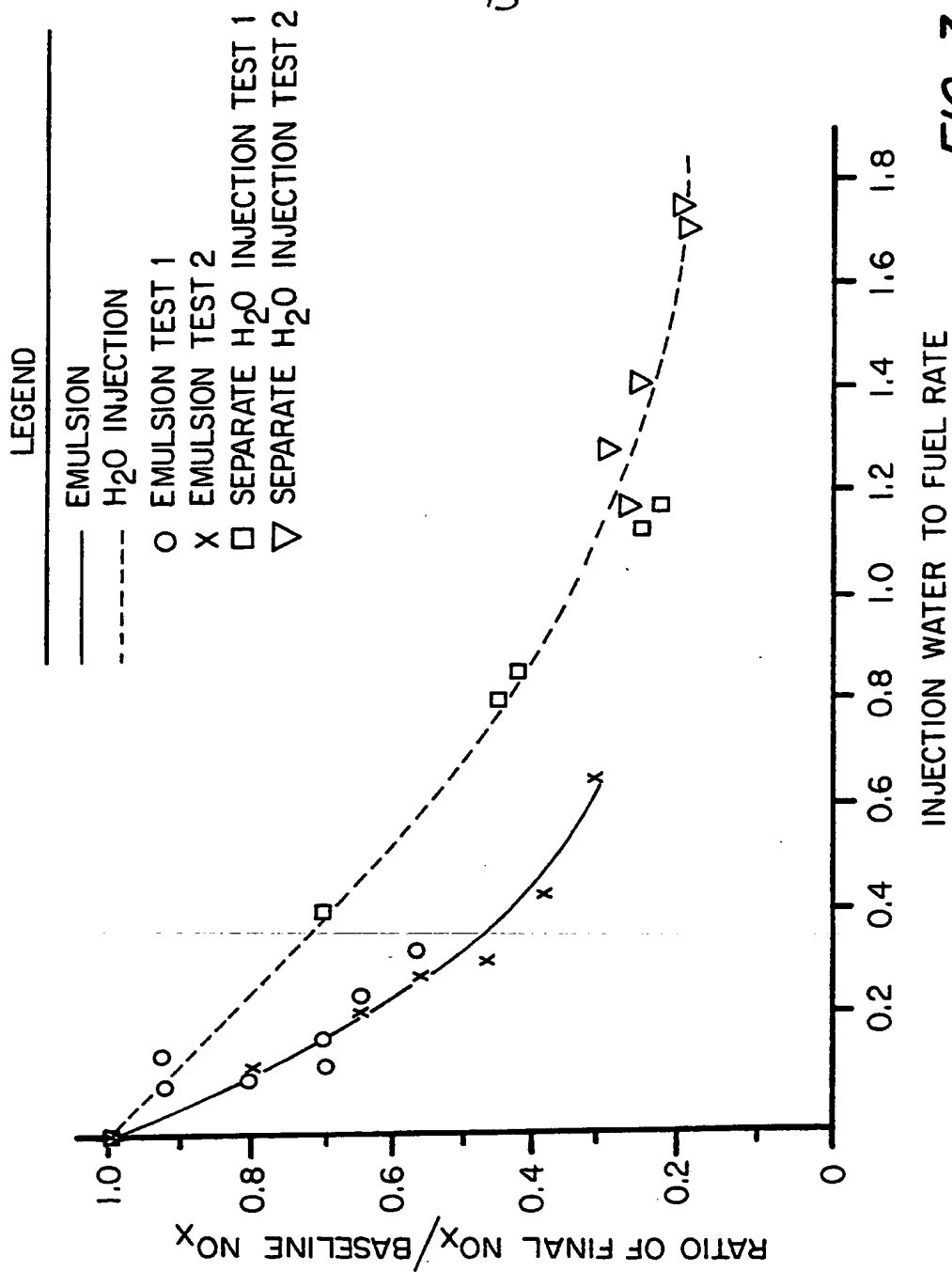


FIG. 3

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INTERNATIONAL SEARCH REPORT

International application No.
PCT/US92/03328

A. CLASSIFICATION OF SUBJECT MATTER

IPC(5) : C10L 1/22

US CL :

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 44/300,418

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

None

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A, 2,892,694 (Weeks) 30 June 1959 See col. lines 40-51.	1-16
A	US,A, 3,281,438 (Johnson) 25 October 196 see col. 2 and 3.	6-8 and 13-15
Y	US,A, 3,339,145 (Martinek et al) 27 August 1968 see col., lines 57-70 and col.2, lines 9-35.	1-16
A	US,A, 3,490,237 (Lissant) 20 January 1970 see cols. 1-4	1-16
A	US,A, 3,637,357 (Nixon et al) 25 January 1972 see col. 1, line 31-end	1-16
A	US,A, 3,932,476 (Bergeron) 13 January 1976 , see col. 2, lines 15-51	6-8 and 13-15
A	US,A, 4,017,522 (Bailey et al) 12 April 1977 see Table 1 of cols 3 and 4	6-8 and 15
A	US,A, 4,083,698 (Wenzel et al) 11 April 1978 see claims 1-5 and 14	1-16
A	US,A, 4,162,143 (Yount III) 24 July 1979 see cols 1 and 2	1-16

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be part of particular relevance	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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Date of the actual completion of the international search 07 JULY 1992	Date of mailing of the international search report 03 AUG 1992
Name and mailing address of the ISA/ Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703)308-3718	Authorized officer <i>Margaret B. Medley</i> MARGARET B. MEDLEY Telephone No. (703) 308-2518

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US92/03328

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
<u>X</u> Y	US,A, 4,173,455 (Fod or et al) 6 November 1979 see claim 1 and col. 2 lines 18-20	<u>1 and 4</u> 2,3 and 5-16
Y	US,A, 4,182,614 (Moriyma et al) 8 January 1980 see col. 2 lines 50 to col. 3 lines 1-31	1-6
A	US,A, 4,199,326 (Fung) 22 April 1980 see col. 1 lines 31 to col. 2, lines 1-47	1-6 and 10-13
<u>X</u> Y	US,A, 4,297,107 (Boehmke) 27 October 1981 see col. 1 lines 2-6 and 58 to col. 2 lines 1-35; and col. 4, lines 53-62; and claims 1 and 4	<u>1,4-5 and 7</u> 2,3,
A	US,A, 4,378,230 (Rhee) 29 March 1983 see col. 2, lines 24-end	1-4, 9-11 and 16
A	US,A, 4,392,865 (Grosse et al) 12 July 1983 see cols. 2, and 5-7	1-16
A	US,A, 4,666,457 (Hayes et al) 19 May 1987 see claims	1-16
<u>X</u> Y	US,A, 4,696,683 (Dentherder) 29 September 1987 see claims	<u>1-4</u> 5-16
Y	US,A, 4,725,287 (Gregoli et al) 16 February 1988 see col. 13, lines 9-16 and 61 to col. 14, lines 1-40 and col. 18, lines 31-51	1-16
A	US,A 4,842,616 (Verhille) 27 June 1989 see col. 1, lines 34 end	1-16
A	US,A, 4,907,368 (Mullay et al) 13 March 1990 see col. 4, lines 23-54	1-16
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A	Scher Chem LS, INC., Technical Bulletin #331-1 August 1983 "Schercomid ODA"	6-8 and 13-15

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